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Author: BARRETT FOUNTOS at EH-07

te: 1/27/98 4:00 PM

riority: Normal

TO: SUE ANDERSON

TO: TOM BELL

TO: MOHANDAS BHAT

TO: FRANK HAWKINS

TO: RUTH NETA

TO: EDWARD PODOLAK

TO: JOSEPH WEISS

TO: ELIZABETH WHITE

TO: JOY WILSON at NOTES2CC

TO: dtripp@bitmailer.net at INTERNET

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TO: Richmondcr@aol.com at INTERNET

TO: henri.metivier@ipsn.fr at INTERNET

TO: sharpl@orau.gov at INTERNET

Subject: next palomares publication

----- Forwarded -----

Author: Asuncion Espinosa Canal <asuncion@ciemat.es> at INTERNET 1/27/98  
11:21 AM

TO: BARRETT FOUNTOS at EH-07

Subject: next palomares publication

----- Message Contents -----

Dear Bar: I send you our next publication in Radiation Protection  
Dosimetry. I hope that we will see next march.

Best regards. Asuncion

ASSESSMENT OF DOSES TO ADULT MEMBERS OF THE PUBLIC  
IN PALOMARES FROM INHALATION OF PLUTONIUM AND AMERICIUM

A Espinosa<sup>\*</sup>, A Aragon<sup>\*</sup>, N Stradling<sup>\*\*</sup>, A Hodgson<sup>\*\*</sup> and A Birchall<sup>\*\*</sup>

<sup>\*</sup> CIEMAT - Spain    <sup>\*\*</sup> NRPB - UK

**Abstract**

This paper reappraises the doses to adults in Palomares resulting from an aviation accident in 1966. For this purpose, site- and material-specific data on airborne contamination and the absorption biokinetics of <sup>239</sup>Pu in animals are used in conjunction with recent ICRP biokinetic models. The committed effective doses for members of the public ranging from 0.037 to 0.21 mSv depending on different work practices are well below the limit of 1 mSv y<sup>-1</sup> currently recommended by ICRP. The predicted urinary excretion rates of <sup>239</sup>Pu show that fission track or mass spectrometry techniques must be used for bioassay for cases of low chronic intake.

## Introduction

As a consequence of the aviation accident in the airspace over Palomares, Spain, in 1966, members of the public living in the area have been, and continue to be potentially exposed to  $^{239}\text{Pu}$  (and  $^{241}\text{Am}$ ) by inhalation. Since the accident, comprehensive studies have been undertaken on the airborne concentrations of  $^{239}\text{Pu}$  in urban and agricultural areas<sup>(1)</sup>. More recently the particle size distribution of the aerosols have been measured<sup>(2,3)</sup> and the biokinetics of  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  in the respirable fraction of contaminated soil investigated in laboratory animals<sup>(4)</sup>. These data have been used in conjunction with the human respiratory tract model<sup>(5)</sup> and systemic model for  $^{239}\text{Pu}$ <sup>(6)</sup> recommended by the International Commission on Radiological Protection (ICRP) to calculate the committed effective doses for members of the public. In addition, the predicted urinary excretion rate of plutonium using this protocol have been compared with measured values. This paper summarises the progress to date on a collaborative programme of work between CIEMAT and NRPB under the auspices of the European Union.

## Materials and Methods

The protocol for assessing doses to members of the public and predicting their urinary excretion is shown in Figure 1.

Information on airborne concentrations over a 30y period at different locations in the Palomares area are summarised in Figure 2. These data represent the average annual concentrations based on weekly sample collections of dust below a particle size of  $10\text{ }\mu\text{m}$ . More detailed information is reported elsewhere<sup>(1)</sup>. It is assumed that the dust was inhaled chronically. Particle size distribution measurements made between 1993 and 1997 are summarised in Table 1. The values for the activity median aerodynamic diameter (AMAD) in the urban area and surrounding agricultural region of Palomares was  $2.8\text{ }\mu\text{m}$  ( $\sigma_g 2.7$ ) and  $1.3\text{ }\mu\text{m}$  ( $\sigma_g 3.6$ )<sup>(3)</sup>. Due to the lack of previous data, it is assumed that these values were typical of the aerosols inhaled by the residents of Palomares after the major countermeasures, to reduce environmental contamination, were completed in 1966.

In Palomares, residents may work in the urban or in the urban and agricultural areas. Hence exposure to aerosols of different particle size and different exercise levels associated with the various work practices need to be considered. The values appropriate for the residents are shown in Table 2. It is assumed that the ventilation rates for each exercise level are the same as the reference values recommended by ICRP<sup>(5)</sup>. The

regional deposition patterns in the respiratory tract are given in Table 3. For comparison the default values for an aerosol of 1  $\mu\text{m}$  AMAD<sup>(5)</sup> are included.

After inhalation of the dust it is assumed that the clearance kinetics by particle transport are the same as in the respiratory tract model<sup>(5)</sup>. The values for the absorption parameters from lungs into blood (Table 4) were obtained from animal studies after administration of the respirable fraction of contaminated Palomares soil<sup>(4)</sup>.

At CIEMAT, measurements of  $^{239}\text{Pu}$  in urine have been made for 30y by alpha spectrometry<sup>(7)</sup>. The detection limits of  $^{239}\text{Pu}$ , by this measuring technique, before and after 1990 were 0.74 and 0.34  $\text{Bq d}^{-1}$  respectively. A small number of measurements have also been made by fission track analysis at the Brookhaven National Laboratory; the detection limit was 2  $\mu\text{Bq d}^{-1}$ . Currently measurements are being made by mass spectrometry and the data will be available shortly.

## Results

The committed effective doses to adults from intakes of  $^{239}\text{Pu}$  during each year of exposure follow the same trend as the airborne concentrations shown in Figure 2. The maximum value, calculated for agricultural workers in 1967, was 54  $\mu\text{Sv}$ . The cumulative values for the committed effective dose arising from intakes over the past 30 years are given in Figure 3. These doses do not include the contributions from  $^{241}\text{Am}$ . Whilst this would have been zero at the time of the accident, the current  $^{239}\text{Pu}$  to  $^{241}\text{Am}$  ratio is about 10:1 by activity. Hence the committed effective doses from intakes of  $^{241}\text{Am}$  since 1966 would at most be 10% of the values in Figure 3 and the total dose increased only marginally.

The predicted daily excretion of  $^{239}\text{Pu}$  at the end of each year after the accident is shown in Figure 4. The data obtained from measurements made by fission track analysis are given in Table 5 and represent the only available urinary excretion data relating to chronic exposure. Clearly these results include the contribution from fall-out due to the testing of nuclear weapons. No information is available for Spain but for the residents of Utah and Colorado (USA), the mean values for persons born before 1958 and after 1961 were 0.6  $\mu\text{Bq l}^{-1}$  (range 0-3.4  $\mu\text{Bq l}^{-1}$ ) and 0.3  $\mu\text{Bq l}^{-1}$  equivalent to about 0.8 and 0.4  $\mu\text{Bq d}^{-1}$ . The contribution to the total dose and urinary excretion from the ingestion of potentially contaminated foodstuffs at Palomares will be taken into account later in the collaborative program between CIEMAT and NRPB.

## Discussion

The assessments of doses for the residents of the Palomares area described in this paper have used field measurements of airborne concentration and particle size, and material specific information on absorption from lungs into blood in conjunction with the most recent biokinetic models recommended by ICRP. The calculated committed effective doses arising from the total intake over 30 years, 37 and 210  $\mu\text{Sv}$  for urban and agricultural workers respectively, are below the values recommended by ICRP, 1  $\text{mSv y}^{-1}$  for members of the public<sup>(9)</sup>.

It is noteworthy that the predicted urinary excretion rates for  $^{239}\text{Pu}$  are similar to the measured values. The urinary excretion data support the use of fission track and mass spectrometric methods of analysis. Clearly alpha spectrometry with a detection limit of 0.34  $\text{mBq d}^{-1}$  would only be of value for assessing doses appreciably in excess of the recommended limit.

## Acknowledgements

We wish to express our appreciation to Dr E. Kaplan for the help in the fission track analysis at the Brookhaven National Laboratory (USA), and to ENRESA and the CEC for the partial financial support necessary to carry out this work.

## References

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**Table 1. Particle size measurements of airborne dust at Palomares**

Date	Urban area		Rural area	
	AMAD $\mu\text{m}$	$\sigma_g$	AMAD $\mu\text{m}$	$\sigma_g$
22/6 to 25/6/93			1.7	3.4
28/6 to 30/6/93			1.3	3.6
20/10 to 4/11/95	3.0	2.7		
17/4 to 22/4/97	2.6	2.4		
17/4 to 28/4/97			0.9	4.8

**Table 2 Daily percentage time budget for adult workers**

Exercise level	Ventilation rates <sup>(5)</sup> (m <sup>3</sup> x h <sup>-1</sup> )	Rural Worker <sup>a</sup> Daily Time budget (%)		Urban Worker <sup>b</sup> Daily Time budget (%)	ICRP-71 Default values (%)
		Urban area	Rural area		
Sleep	0.45	33 <sup>c</sup>	0 <sup>c</sup>	34 <sup>c</sup>	33,3
Sitting	0.54	32 <sup>c</sup>	0 <sup>c</sup>	36 <sup>c</sup>	25,0
Light exercise	1.50	12 <sup>c</sup>	5 <sup>c</sup>	20 <sup>c</sup>	40,6
Heavy exercise	3.00	3 <sup>c</sup>	15 <sup>c</sup>	10 <sup>c</sup>	1,0

<sup>a</sup> Inhabitants of Palomares participating in agricultural labors at the contaminated site

<sup>b</sup> Inhabitants of Palomares living and working at the urban area

<sup>c</sup> Direct estimations from local data.



**Table 3 Deposition of aerosols in respiratory tract**

Region	% inhaled activity		ICRP <sup>b</sup>
	Urban worker <sup>a</sup>	Rural worker <sup>a</sup>	
ET <sub>1</sub>	22.05	17.79	14.89
ET <sub>2</sub>	30.77	25.53	18.97
BB	4.20	4.13	1.29
bb	2.21	2.20	1.95
AI	10.24	10.87	11.48
Total	69.47	60.53	48.58

<sup>a</sup> Values calculated using AMADs and time budget given in Tables 1 and 2 and ICRP default values for ventilation rates.

<sup>b</sup> Default values recommended by ICRP for an aerosol of 1  $\mu\text{m}$  AMAD<sup>(5)</sup>

**Table 4 Absorption parameter values for  $^{239}\text{Pu}$  and  $^{241}\text{Am}$**

Parameters	Pu	Am	Type M	Type S
$f_r 10^{-2}$	4.2	8.2	10	0.1
$s_r \text{ d}^{-1}$	100	100	100	100
$s_s 10^{-4} \text{ d}^{-1}$	2.0	3.9	50	1

$f_r$ ,  $s_r$  and  $s_s$  are the rapidly dissolved fraction, rapid and slow dissolution rate respectively<sup>(5)</sup>.

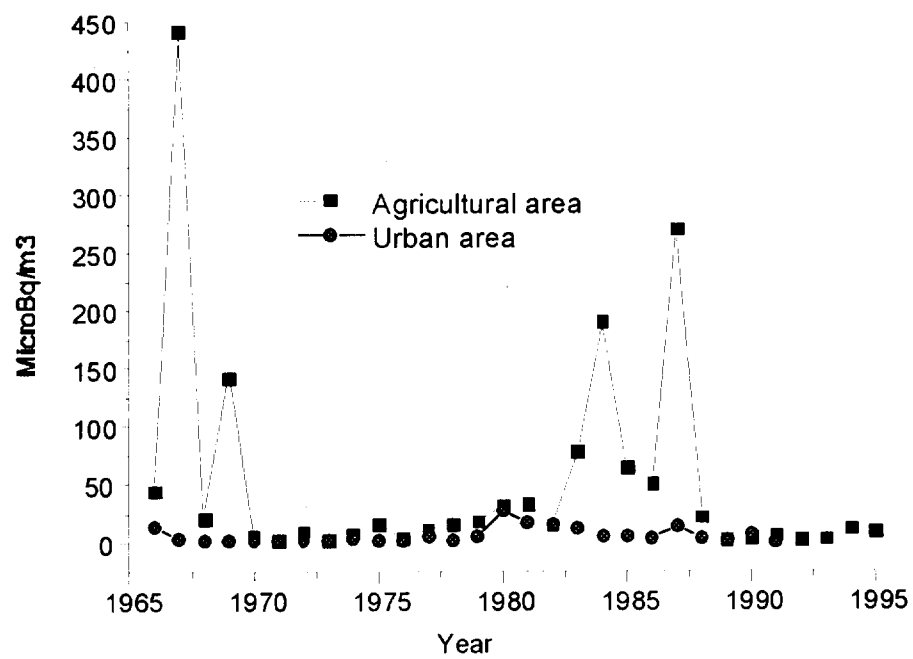
Values of the model parameters  $s_p$ ,  $s_{pt}$  and  $s_t$  can be calculated readily from them<sup>(4)</sup>. In the absence of data during the early lung clearance phase the default value of  $100 \text{ d}^{-1}$  for  $s_r$  was assumed.

**Table 5 Daily urinary excretion data for man working in agricultural area**

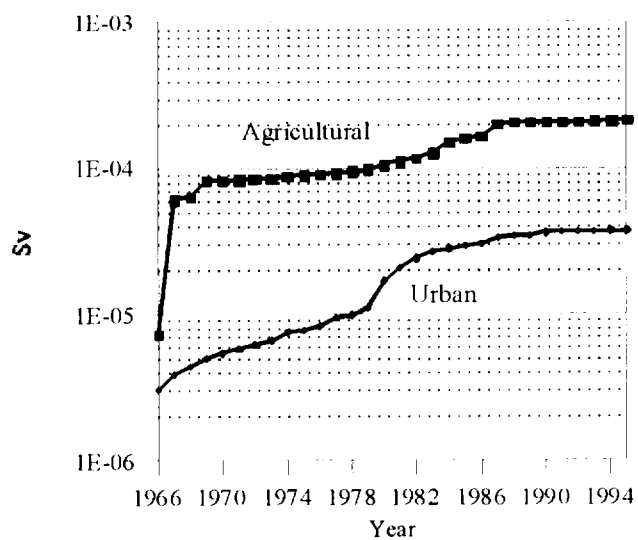
Date	$^{239}\text{Pu}$ ( $\mu\text{Bqd}^{-1} \pm 3\sigma$ ) Fission track method
25/7/93	$10.6 \pm 2.4$
26/7/93	$12.8 \pm 1.2$
27/7/93	$20.8 \pm 4.3$
28/7/93	$27.8 \pm 3.7$

**Figure 1. Protocol for assessing doses from inhaled dust at Palomares**

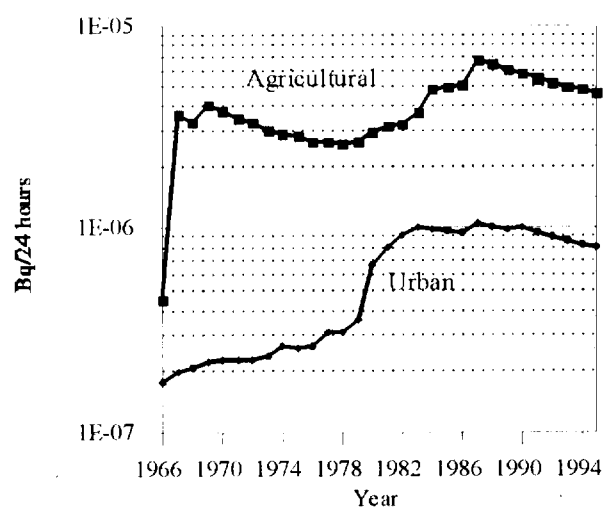
**Figure 2. Airborne concentration of  $^{239}\text{Pu}$  in Palomares.**



**Figure 3**    **Committed effective doses**



**Figure 4** Predicted daily excretion of  $^{239}\text{Pu}$



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Author: BARRETT FOUNTOS at EH-07

Date: 1/27/98 4:24 PM

Priority: Normal

Receipt Requested

TO: FRANK HAWKINS

TO: EDWARD PODOLAK

Subject: Resignation of Fred Mettler

----- Message Contents -----

This afternoon, Fred Mettler informed Linda Sharp of ORISE that he decided to withdraw as a panelist from the Palomares Program Review. He cited personal reasons -- his son is in a collegiate swim meet the weekend we planned to leave for Spain.

I will begin searching for a replacement ASAP.